# PATENT SPECIFICATION

(11) 1 485 694

10

15

20

25

30

35

(21) Application No. 2873/75 (22) Filed 22 Jan. 1975

(44) Complete Specification published 14 Sept. 1977

(51) INT CL<sup>2</sup> B01F 17/28; A61K 7/06, 7/48; C09D 5/04; C10M 1/36, 1/38

(52) Index at acceptance

2A3 2AX 2B 2D 2E 3DX B<sub>1</sub>V

C14B C17 C20A C20D1 D1A T2E

27C C11 C12 C16 C29 C4 C9A L2A L4G C3R

6F2 6H2 C3T

9B4 9BX 9E2

C5F 600 646 647 808 C

C5G



5

10

15

20

25

30

35

(72) Inventors TADAOMI SAITO, YOSHIMASA MATSUZAWA, SADAYOSHI NINAGAWA, MASAO HONMA, MASAHIKO TAKESADA and MASAHIRO TAKEHARA

#### (54) GELLING AGENTS

(71) We, AJINOMOTO CO., INC., a corporation organised under the laws of Japan, of No. 6, 1-chome, Kyobashi, Chuo-ku, Tokyo, Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:-

This invention relates to the gelation or solidification of non-polar organic

liquids, and to the products thereby obtained.

Non-polar organic liquids are used for various purposes, and the more common thereof include fuels, motor oils, paints, cosmetics and edible oils.

When using non-polar organic liquids, it is often desirable to place them in a

solid state. When a container containing such liquids is damaged or broken open, the organic liquids stored therein otherwise flow out and spread over a wide area and their recovery is difficult. If the organic liquids which have flowed out are inflammable, they may cause a fire by spontaneous ignition. Fires and smoke inhalation are often causes of many deaths in aircraft accidents in which otherwise there would be a reasonable chance of survival. The fires are usually caused when the highly volatile fuel spills from damaged tanks and becomes widely distributed. Fuel vaporizes and is easily ignited by hot engine parts or sparks from metal impact. When fuels are gelled, however, the degree of vaporisation and the extent to which the fuel becomes distributed upon impact are reduced, and so the danger of a rapidly spreading fire or explosion is substantially reduced. Hence, the gelling of fuels has been considered as a safety factor in jet aircraft. Gelled fuels may be handled like fluid liquids when subject to shearing forces. Moreover, gelling agents for organic liquids are often useful for facilitating the removal or collection of spilt non-polar organic liquids or mixtures of non-polar organic liquids, and for prevention of leakage or spills of such non-polar organic liquids from leaking tanks or holes.

It has now been found that certain N-acyl amino acids and derivatives of Nacyl amino acids having a dielectric constant of not more than 20 and a solubility in water of no more than 10% by weight, for example esters, amides and amine salts, are highly effective gelling agents for liquid hydrocarbons and other non-polar organic liquids.

According to one aspect of this invention, there is provided a process for preparing a gel which comprises (A) homogeneously admixing a non-polar organic liquid having a dielectric constant of not more than 20 and having a solubility of not more than 10% by weight in water, with at least one N-acylamino acid or derivative thereof employed in an amount sufficient to gel the non-polar organic liquid, N-acylamino acid or derivative thereof possessing the general formula:

15

20

30

35

40

wherein RCO is an aliphatic acyl radical containing from 2 to 30 carbon atoms or an aralkyl acyl radical; Z is a group of formula -OH, -OR, -NH2, -NHR2,



or —O-R<sub>5</sub>NH<sub>3</sub>+, wherein R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub> and R<sub>5</sub> are aliphatic hydrocarbyl radicals containing from 1 to 30 carbon atoms or aralkyl radicals: X is hydrogen or a methyl radical, and Y is a group of formula

---CH---

wherein R<sub>6</sub> is an alkyl radical containing from 1 to 4 carbon atoms, a benzyl radical, a phenyl radical, or a radical of formula -CH2OH,

OH

-CHCH<sub>3</sub>,

10

 $-CH_2SH$ ,  $-CH_2CH_2S-CH_3$ ,  $-(CH_2)_1-COZ$ , wherein 1 is 1 or 2 and Z has the aforesaid meaning,  $-(CH_2)_mNHCOR$  wherein m is 3 or 4 and RCO is as above, or  $-(CH_2)_m$ , wherein n is an integer of from 1 to 6, with the proviso that when Z is  $-OR_1$ , Y is

15

wherein R<sub>6</sub> is --(CH<sub>2</sub>)<sub>m</sub>NHCOR and that at least one of R, R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub> and R<sub>5</sub> is an aliphatic hydrocarbyl radical containing from 5 to 30 carbon atoms.

and (B) allowing the mixture to stand until gel formation occurs.

This invention also provides a gel which comprises a homogenous mixture

20

25

30

of (a) a non-polar organic liquid having a dielectric constant of not more than 20 and a solubility in water of not more than 10°, by weight and (b). present in an amount sufficient to gel the non-polar organic liquid, at least on N-acylamino acid or derivative thereof having the general formula:

wherein RCO, X, Y and Z have the meanings set out above 25 In the above general formula, the RCO radical is an aliphatic acyl radical derived from an alkanoic or alkenoic acid containing from 2 to 30 carbon atoms.

for example, acetic acid, propionic acid, butyric acid, caproic acid, caprylic acid, capric acid, lauric acid, myristic acid, palmitic acid, stearic acid, oleic acid, linoleic acid, elaidic acid, naturally occurring mixtures of such acids, for example coconut oil fatty acid, tallow oil fatty acid and hydrogenated tallow oil fatty acid

and synthetic fatty acids. The RCO radical can also be an aralkyl acyl radical, for

example phenylacetyl,  $\alpha$ -naphthaleneacetyl or hydrocinnamoyl.

R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub> and R<sub>5</sub> are alkyl or alkenyl radicals containing from 1 to 30 carbon atoms, for example methyl, ethyl, propyl, butyl, amyl, hexyl, cyclohexyl, octyl, nonyl, decyl, dodecyl, undecenyl, undecyl, tridecyl, tetradecyl, cetyl, pentadecyl, heptadecyl, hexadecyl, hexadecenyl, octadecyl, octadecenyl, octadecadieyl, oleyl, and elaidyl, or are aralkyl radicals for example benzyl and

hydrocinnamyl. The amino acid moiety of a gelling agent comprised by a composition 40

35

according to this invention, i.e. a moiety of general formula:

BNSDOCID: <GB\_\_\_1485694A\_\_I\_>



is preferably derived from an acidic amino acid, a neutral amino acid, a sulphur-containing amino acid, a hydroxy amino acid, a N-methyl derivative of an  $\alpha$ -amino acid, an  $\omega$ -amide of an acidic amino acid, a basic amino acid, a  $\beta$ -amino acid or an 5  $\omega$ -amino acid. 5 The amino acid moiety is particularly preferably derived from one of the following amino acids: aspartic acid, glutamic acid, glutamine, glycine, sarcosine,  $\alpha$ -alanine,  $\beta$ -alanine,  $\alpha$ -aminobutyric acid, valine, norvaline, leucine, isoleucine, norleucine, phenylglycine, phenylalanine, serine, threonine, cysteine, methionine,  $N^s$ -acylornitine,  $N^s$ -acyllysine,  $\gamma$ -aminovaleric acid and  $\omega$ -aminocaproic acid. 10 At least one aliphatic hydrocarbyl radical containing from 5 to 30 carbons 10 must be present in the gelling agents used in the practice of this invention, either in the N-acyl moiety or in the other constituent moiety, i.e. ester, amide or amine salt. Both moieties may include such aliphatic hydrocarbyl radical, if desired. Specific examples of gelling agents which may be used in gel compositions 15 15 embodying this invention include the following compounds: (1) N-acyl amino acids: N-lauroyl-αalanine; N°, N°-dicaproyl-, N°, N°-dicaproyl-, N°, N°-dicaproyl-, N°, N°-dilauroyl- and N°, N°-dicaproyl-ornithine (cocoyl indicates the acyl radical of coconut oil fatty acid); N°, N°-dicaproyl- and Na, Ne-dicapryloyllysine; N-lauroyl valine and N-lauroyl glutamic acid. 20 (2) N-acyl amino acid esters: N°, N°-dicapryloylornithine octyl, decyl, lauryl and stearyl esters, N°, N°-dicapryloyllysine octyl, decyl and lauryl esters; N°, N°-dicapryloyllysine hexyl, octyl, decyl and lauryl esters; N°, N°-dicocoyllysine hexyl, 20 octyl, decyl and lauryl esters; and No, No-di(tallowyl) and No, No-di(hydrogenated tallowyl) lysine hexyl, octyl, decyl and lauryl esters (tallowyl indicates the acyl 25 radical of tallow oil fatty acid). From the identified esters, it will be appreciated 25 that the alkyl esterifying groups preferably contain from 6 to 18 carbon atoms. Other N-acyl amino acid esters which can be used are N°, N³-dilauroyl- N°, N³-distearoyl and N°, N³-ditallowyl-ornithine alkyl esters and N°, N¹-distearoyl-lysine alkyl esters. 30 (3) N-acylamino acid amides: N-acetyl glutamic acid  $-\alpha, \gamma$ -diamides wherein 30 the alkyl radicals have 12 to 18 carbon atoms, for example N-acyl glutamic acid the alkyl radicals have 12 to 16 carbon atoms, for example 14-acyl glutamic acid  $\alpha, \gamma$ -dilauryl or  $-\alpha, \gamma$ -disearyl amide; N-caproyl glutamic acid  $\alpha, \gamma$ -diamide and N-caproyl glutamic acid $\alpha, \gamma$ -dialkylamides wherein the alkyl radicals have 1 to 18 carbon atoms, for example N-caproyl glutamic acid  $-\alpha, \gamma$ -dibutyl,  $-\alpha, \gamma$ -dihexyl,  $-\alpha, \gamma$ -dibutyl,  $-\alpha, \gamma$ -di 35 35  $\alpha, \gamma$ -dioctyl,  $-\alpha, \gamma$ -dilauryl and  $-\alpha, \gamma$ -distearyl amides; N-lauroyl glutamic acid  $-\alpha, \gamma$ -diamide and N-lauroyl glutamic acid $\alpha, \gamma$ -dialkylamides wherein the alkyl radicals have 1 to 18 carbon atoms, for example N-lauroyl-glutamic acid  $-\alpha, p$ -dibutyl,  $-\alpha, p$ dihexyl,  $-\alpha, p$ -dioctyl,  $-\alpha, p$ -dilauryl and  $-\alpha, p$ -distearyl amides; N-cocoyl glutamic acid  $-\alpha, p$ -diamide and N-cocoyl glutamic acid  $-\alpha, p$ -diamide and N-cocoyl glutamic acid  $-\alpha, p$ -diamide and N-cocoyl glutamic acid  $-\alpha, p$ -diamides wherein the alkyl radicals have 1 to 18 carbon atoms, for example N-cocoyl glutamic acid  $-\alpha, p$ -dibutyl,  $-\alpha, p$ -dioctyl,  $-\alpha, p$ -dilauryl and  $-\alpha, p$ -diamides; N-(tallowyl or hydrogenated tallowyl) glutamic acid  $-\alpha, p$ -diamide and N—(tallowyl or hydrogenated tallowyl)  $-\alpha, p$ -dialkylamides wherein the alkyl radicals have from 1 to 18 carbon atoms. for example N-(tallowyl or hydrogenated tallowyl)  $-\alpha, p$ -dialkylamides wherein the alkyl radicals have from 1 40 40 to 18 carbon atoms, for example N-(tallowyl or hydrogenated tallowyl)  $-\alpha$ ,  $\gamma$ -45 dibutyl,  $-\alpha,p$ -dihexyl,  $-\alpha,p$ -dioctyl,  $-\alpha,p$ -dilauryl and  $-\alpha,p$ -distearyl amides;  $N^{\alpha},N^{\epsilon}$ -diacetyllysine octyl, lauryl and stearyl amides;  $N^{\alpha},N^{\epsilon}$ -dicaproyllysine amide and  $N^{\alpha},N^{\epsilon}$ -dicaproyllysine alkyl amides wherein the alkyl radicals contain from 1 to 18 45 carbon atoms, for example N°, N'-dicaproyllysine butyl, hexyl, octyl, lauryl and stearyl amides; N°, N'-dicaproyllysine amide and N°, N'-dicaproyllysine alkyl 50 amides wherein the alkyl radicals contain from 1 to 18 carbon atoms, for example 50 amides wherein the alkyl radicals contain from 1 to 18 carbon atoms, for example N°,N'-dicaproyllysine, butyl, dibutyl, hexyl, octyl, lauryl and stearyl amides; N°,N'-dilauroyllysine amide and N°,N'-dilauroyllysine alkyl amides wherein the alkyl radicals contain from 1 to 18 carbon atoms, for example N°,N'-dilauroyllysine butyl, dibutyl hexyl, octyl, lauryl and stearyl amides; N°,N'-dicocoyllysine amide, and N°,N'-dicocoyllysine alkyl amides wherein the alkyl radicals contain from 1 to 18 carbon atoms, for example N°,N'-dicocoyllysine butyl, dibutyl, hexyl, octyl, lauryl, and stearyl amides, N°,N'-dichydrogenated tallowyll, dibutyl, hexyl, octyl, lauryl, and stearyl amides, N°,N'-dilhydrogenated tallowyll, 55 55 dibutyl, hexyl, octyl, lauryl and stearyl amides,  $N^{\alpha}$ ,  $N^{\epsilon}$ -di(hydrogenated tallowyl) lysine amide, and  $N^{\alpha}$ ,  $N^{\epsilon}$ -di(tallowyl or hydrogenated tallowyl) lysine alkyl amides wherein the alkyl radicals contain from 1 to 18 carbon atoms, for example Na, No-60 60

di(tallowyl or hydrogenated tallowyl) lysine butyl, dibutyl, hexyl, octyl, lauryl and stearyl amides. N-lauroylvaline amide and butyl, hexyl, octyl and lauryl amides. It

| 4  | 1,465,07   |      |
|----|--|------|
|    | is also possible to use N-stearoyl glutamic acid $-\alpha, \gamma$ -diamide, N-stearoyl glutamic acid $-\alpha, \gamma$ -dialkylamides wherein the alkyl radicals have 1 to 18 carbon atoms, N°, N°-distearoyllysine amide and N°, N°-distearoyl-lysine alkyl amides wherein the   |      |
| 5  | alkyl radical has 1 to 18 carbon atoms, and, for example, relations in successful amide and butyl, hexyl, octyl and lauryl amides; N-capryloyl leucine amide and butyl, hexyl, octyl and lauryl amides; and N-palmitoylmethionine amide and butyl,   | 5    |
|    | hexyl and octyl amides.  (4) A N-acyl amino acid amine salt may be a N°,N'-dicaproyllysine alkyl-  | 10   |
| 10 | N°, N'-dicocoyllysine alkylamine salt wherein the alkylamine has 4 to 18 carbon atoms, a N°, N'-disearcyllysine alkylamine salt wherein the alkylamine has 4 to 18 atoms, a N°, N'-disearcyllysine alkylamine salt wherein the alkylamine has 4 to 18  | (    |
| 15 | wherein the alkylamine has 4 to 18 carbon atoms, a 1°, 1° discarbon atoms, a N°, N°-alkylamine salt wherein the alkylamine has 8 to 18 carbon atoms, a N°, N°-alkylamine has 4 to 18 carbon  | 15 ' |
|    | atoms, a N°, N°-dicocoylornithine alkylamine salt wherein the alkyl-<br>18 carbon atoms, a N°, N³-distearoylornithine alkylamine salt wherein the alkyl-   | 20   |
| 20 | wherein the alkylamine has 4 to 18 carbon atoms, N-lauroyl-phenylalanine alkylamine salt wherein the alkylamine has 8 to 18 carbon atoms, a N-cocoylphenylalanine alkylamine salt wherein the alkylamine has 8 to 18 carbon atoms, a N-stearoylphenylalanine alkylamine salt wherein the alkylamine has 8 to 18 carbon stearoylphenylalanine alkylamine salt wherein the alkylamine has 8  |      |
| 25 | atoms or a N-tallowylphenylalanine alkylanine salt whether the alkylanine salts N°, N°- to 18 carbon atoms. Examples of specific such compounds are N°, N°- to 18 carbon atoms. Examples of specific such compounds are N°, N°-  | 25   |
|    | butyl, octyl, lauryl amine salts, N°,N'-dicocoyllysine butyl, octyl, lauryl and stearyl amine salts, N°,N'-dicocoyllysine butyl, octyl, lauryl and stearyl amine salts, N°,N'-di(hydrogenated tallowyl) lysine butyl, octyl, lauryl and stearyl amine salts, N-lauroylphenylalanine octyl lauryl, and stearyl amine  | 30   |
| 30 | salts.  Gelling agents which may be employed in the compositions of this invention.  can agglomerate to form a coherent structure and trap the non-polar organic   |      |
| 35 | acyl amino acids such as N-acylgiutamic acid diesters and N-acyl heatrai amino acid esters cannot agglomerate to form a coherent structure and so cannot be used   | 35   |
| 40 | Among the four kinds of gelling agents which may be employed in this invention, and which are exemplified by compounds of the four aforesaid groups (1) to (4), N-acyl amino acids may be easily prepared by acylating amino acids with acyl chlorides in aqueous media in the presence of bases, for example sodium hydroxide. Esters or amides of N-acylamino acids may be obtained by reacting N-hydroxide. Setters or amines or amines or ammonia in the presence or absence | 40   |
| 45 | of an acidic catalyst, reaction being accompanied by neutralizing an N-acyl amino acid with an   | 45   |
| 45 | The gel compositions of this invention may be prepared simply by forming a homogeneous mixture of a gelling agent of the above general formula (I) with a non-linear size liquid and then allowing the mixture to stand until gel formation  |      |
| 50 | occurs. Homogeneous admixture may be carried out at temperatures ranging from room temperature to the boiling point of the non-polar organic liquid. A gelling agent may be added in the form of a very fine powder or in solution in suitable solvents, for example acetone, methanol or ethanol, or in a small amount of the hot non-polar organic liquid to be gelled. A gel composition of this invention may  | 50   |
| 55 | temperature and thereafter cooling the resultant solution to a temperature at which gel formation occurs on standing. In all cases gel formation will occur when a mixture of a gelling agent and non-polar organic liquid has become sufficiently   | 5,5  |
| 60 | homogeneous.  The gelling agent will usually be employed in an amount of from 0.05% to 20% by weight of the non-polar organic liquid to be gelled. The firmness of the resultant by weight of the non-polar organic liquid to be gelled. The firmness of the resultant   | 60   |
|    | However, in general, the addition of more than 10% by weight of gening agent causes no appreciable increase in the shear strength of the resultant gel. The  | 65   |
| 65 | polar organic liquid. The required amount of gelling agent may be determined   | 03   |

|    | experimentally and the amount of a particular gelling agent used will vary in accordance with the desired physical properties of the gel and in accordance with the affect on gelling of any substances in solution in the organic liquid.   |    |
|----|--|----|
| 5  | invention are those having a dielectric constant of not more than 20 and having a solubility of not more than 10% by weight in 100 for more than 20 and having a   | 5  |
|    | gasoline, naphthas kerosene gas oil beaut oil and and a little to the sample   |    |
| 10 | example benzene, xylene, toluene hexage heptane octano and available.  | 10 |
|    | phthalate, dioctyl phthalate, diethyl sebacate and dioctyl sebacate; ketones, for example methyl isobutyl ketone and dissobutyl ketone; aldebudge for  | 10 |
| 15 | chloroethylene and chlorobenzene; phosphoric esters for example carbon tetrachloride, tetra-   |    |
|    | brooking and therest bhosphate, hund boltotaleries monocilist c.   | 15 |
|    | polyoxyethylene monostearyl ethers, polyoxypropylene mono C to C all all at least and the control of the contro |    |
| 20 | containing from to to be example polyoveneousland  |    |
| 20 | monobutyl ether, polyoxypropylene monolauryl ether or polyoxypropylene monostearyl ether; liquid polyoxyalkylene glycol fatty acid esters, for example   | 20 |
|    | oxyethylene units; fatty oils, for example corn oil soya bean oil olive oil and oil  |    |
| 25 | on, conton seed on, saiding oil, nerring oil and whale oil and cilicone oil Mivey-   |    |
|    | of these liquids and liquids containing predominantly these non-polar organic liquids can be used.   | 25 |
|    | Organic liquids having a dielectric constant of more than 20, such as acetone and ethanol, and having a solubility of more than 10% by weight in water such as acetic acid and butyric acid have been found and butyric acid have been found and the solution of the solution  |    |
| 30 |  | •  |
|    | The non-polar organic liquids used in compositions embodying this invention may contain other ingedients as may be required to impart special properties thereto. An emulsion or suspension of the required to impart special properties   | 30 |
|    | be gelled. In the case of a system which forms a water-in oil type of a system which   |    |
| 35 | suspension whose water content is below 50%, the entire system may be gelled. In the case of a system which forms an oil-in-water type of emulsion or suspension, it is possible that only the non-polar organical liquid.   |    |
|    | gelled.  | 35 |
| 40 | The process of this invention may be used, in particular, in the gelation of fuel oil and recovering of drained oil. The aforesaid gelling agents may be used in the   |    |
| 40 | agent for paints or inks, a gelling agent for greases, a colidifician asset for  | 40 |
|    |  |    |
| 45 | cosmetics, a binder or carrier for medicaments and a gelling agent for napalm-type combustion charges.   |    |
| 45 | In general, no refined techniques are needed to produce gelled non-polar organic liquids by the process of this invention. The gelling agents used are capable of gelling non-polar organic liquids when used  | 45 |
|    | The second was point of same fidules when likely in small amounts  |    |
| 50 | Moreover, when the gelling agents are employed for fuel oils, fuels are gelled and hence rendered much safer. The gelling agent is itself flammable and exerts no influence on the flammability of the fuel  |    |
| 50 | influence on the flammability of the fuel.  The following examples illustrate this invention:  | 50 |
|    | Example 1.  One gram of each of the N-acylamino acid derivatives set out in Table 1 was dissolved in 100 gram samples of kerosana and of allowed in Table 1 was  |    |
| 55 | dissolved in 100 gram samples of kerosene and of olive oil with stirring at a temperature of 120°C. The solutions obtained were cooled to room temperature.  | 55 |
|    | After standing for 2 hours, gelled kerosene or olive oil formed. The shear strength of these gels was measured with a rheometer, RUD—J type (a product of Fuji Rika Kogyo Co.). The results obtained are also set out in Table 1.  |    |
|    |  |    |

### TABLE 1

| Shear strength of ge  |                   | ngth of gel        |
|---|-------------------|--------------------|
| Derivatives of N-acylamino acids                              | Kerosene<br>g'cm² | Olive oil<br>g/cm² |
| Na,Nc-dicaproyllysine lauryl ester                            | 36                | 122                |
| Na,N <sup>€</sup> -distenroyllysine n-hexyl ester             | 14                | 63                 |
| Nα,Nε-dilauroyllysine benzyl ester                            | 9.3               | 7.6                |
| Na,Ne-di(2-ethyl-hexanoyl)lysine lauryl ester                 | 15                | 16                 |
| Na,No-dicapryloylornithine lauryl ester                       | 13                | 36                 |
| N-lauroylglutamic acid-α,γ-diamide                            | 28                | 29                 |
| N. lauroylglutamic acid-α, γ-di-n-butylamide                  | 223.              | 235                |
| N-lauroylglutamic acid-α,γ-distearylamide                     | 41                | 26                 |
| N-acetylglutamic acid-a,y-dilaurylamide                       | 13                | 25 .               |
| N-phenacetylglutamic acid-α,y-dilaurylamide                   | 11                | 10                 |
| N-lauroylglutamic acid-a,y-di(2-ethyl hexylamide)             | 35                | 28                 |
| N-stearoyl aspartic acid-a, \beta-di-n-butylamide             | 12                | . 13               |
| N-lauroyl valine n-butylamide                                 | 3.0               | 19                 |
| N-lauroyl sarcosine n-butylamide                              | 0.9               | 1.4                |
| N-palm.toyl-e-amino caproic acid n-butylamide                 | 3.0               | 4.0                |
| Na,Ne-dicapryloyllysine laurylamide                           | 153               | 89                 |
| N <sup>a</sup> ,N <sup>e</sup> -dicapryloyllysine benzylamide | 12                | 13                 |
| Laurylamine N-lauroyl valinate                                | 3.2               | 4.1                |
| Stearylamine N-lauroylalaninate                               | 3.1               | 4.2                |
| Laurylamine N-lauroylphenylalaninate                          | 4.0               | 74                 |
| Stearylamine Na, Ne-dicapryloyllysinate                       | 17                | 61                 |
| n-Butylamine Na,N <sup>€</sup> -dilauroyllysinate             | 20                | 25                 |
| Na,Nδ,dicapryloylornithine di-n-butyl amide                   | 1.9               | 2.9                |

In addition to those indicated in Table 1, gelled kerosenes having shear strength values of more than 15 g/cm² were obtained using the following derivatives of N-acylamino acids.

5

derivatives of N-acylamino acids.  $N^{\alpha}, N^{\delta}$ -dicaproyl-,  $N^{\alpha}, N^{\delta}$ -dicaproyl-,  $N^{\alpha}, N^{\delta}$ -dicaproyl-,  $N^{\alpha}, N^{\delta}$ -dicaproyl-,  $N^{\alpha}, N^{\epsilon}$ -dicaproyl-,  $N^{\alpha}, N^{\epsilon}$ -dicaproyl-,  $N^{\alpha}, N^{\epsilon}$ -dicaproyl-,  $N^{\alpha}, N^{\epsilon}$ -dicaproylog ornithine n-octyl, 2-ethylhexyl and decyl esters:  $N^{\alpha}, N^{\delta}$ -dicaproylornithine n-hexyl, n-octyl, 2-ethylhexyl, decyl and lauryl esters:  $N^{\alpha}, N^{\delta}$ -dicocoylornithine n-hexyl, n-octyl, 2-ethylhexyl, decyl and lauryl esters:  $N^{\alpha}, N^{\delta}$ -distearoylornithine n-hexyl, n-octyl, 2-ethylhexyl, decyl and lauryl esters:  $N^{\alpha}, N^{\delta}$ -ditallowylornithine n-hexyl, n-octyl, 2-ethylhexyl, decyl and lauryl esters:

5

10

10

|            |  | ,  |
|------------|--|----|
|            | N <sup>\alpha</sup> , N'-dilauroyllysine n-hexyl, n-octyl, lauryl, 2-ethylhexyl and decyl esters; N <sup>\alpha</sup> , N'-dicocoyllysine n-hexyl, n-octyl, lauryl, 2-ethylhexyl and decyl esters; N <sup>\alpha</sup> , N'-distearoyllysine n-octyl, lauryl, n-ethylhexyl and decyl esters; N <sup>\alpha</sup> , N'-ditallowyllysine n-hexyl, n-octyl, lauryl, 2-ethylhexyl and decyl esters;  |    |
| 5          | N-capryloylglutamic acid-\alpha,p-distearylamide; N-capryloylglutamic acid-\alpha,p-diamide and di-n-butyl, diisobutyl, di-n-octyl, di-2-ethylhexyl, dilauryl distearylamides.   | 5  |
| 10         | N-cocoylglutamic acid-\(\alpha_1\); diamide and di-n-butyl, diisobutyl, di-n-octyl, di-2-ethylhexyl, dilauryl and distearylamides;  N-stearoylglutamic acid-\(\alpha_1\); diamide and di-n-butyl, diisobutyl, di-n-octyl,  | 10 |
|            | N-tallowylglutamic acid- $\alpha$ , p-diamide, di-n-butyl, diisobutyl, di-n-octyl, di-2-ethylhexyl, dilauryl and distearylamide:   | 10 |
| 15         | N-lauroylglutamic acid-\(\alpha_i\), diisobutyl and dilaurylamides; \(N^\alpha_i\), N'-diacetyllysine lauryl and stearylamides; \(N^\alpha_i\), N'-dicapryloyllysine amide and n-butyl, isobutyl, n-octyl, 2-ethylhexyl and stearylamides;   | 15 |
| 20         | N°, N'-dilauroyllysine amide and n-butyl, isobutyl, n-octyl, 2-ethylhexyl, lauryl and stearyl amides:  | •  |
| 20         | N <sup>a</sup> ,N <sup>c</sup> -dicocoyllysine amide and n-butyl, isobutyl, n-octyl, 2-ethylhexyl, lauryl and stearylamides; N <sup>a</sup> ,N <sup>c</sup> -distearoyllysine amide and n-butyl, isobutyl, n-octyl, 2-ethylhexyl, lauryl and stearylamides;  | 20 |
| 25         | N <sup>a</sup> ,N <sup>c</sup> -ditallowyllsine amide and n-butyl, isobutyl, n-octyl, 2-ethylhexyl, lauryl and stearylamides:  | 25 |
|            | N°.N°-dicapryloyllysine n-octyl-, 2-ethylhexyl- and lauryl-amine salts;<br>N°,N°-dilauroyllysine n-octyl-, 2-ethylhexyl-, lauryl- and stearyl amine salts;<br>N°,N°-dicocoyllysine n-butyl-, isobutyl-, n-octyl-, 2-ethylhexyl-, lauryl- and stearyl amine salts;  | 20 |
| 30         | $N^{\alpha}$ , $N^{\epsilon}$ -distearoyllysine n-butyl-, isobutyl-, n-octyl-, 2-ethylhexyl-, lauryl- and stearyl amine salts; $N^{\alpha}$ , $N^{\epsilon}$ -ditallowyllsine n-butyl, isobutyl-, n-octyl-, 2-ethylhexyl-, lauryl-, and  | 30 |
| 35         | stearyl amine salts;  N <sup>a</sup> ,N <sup>b</sup> -dicapryloylornithine n-octyl-, 2-ethylhexyl-, lauryl- and stearyl amine salts:   |    |
| 33         | N°, N°-dilauroylornithine n-butyl-, isobutyl-, n-octyl-, 2-ethylhexyl-, lauryl- and stearyl amide salts; N°, N°-dicocoylornithine n-butyl-, isobutyl-, n-octyl-, 2-ethylhexyl-, lauryl-, n-octyl-, n-oct | 35 |
| 40         | N <sup>a</sup> , N <sup>b</sup> -distearoylornithine n-butyl-, isobutyl-, n-octyl-, 2-ethylhexyl-, lauryl- and stearyl amine salts:  | 40 |
|            | $N^{\alpha}, N^{\delta}$ -ditallowylornithine n-butyl-, isobutyl-, n-octyl-, 2-ethylhexyl-, lauryl- and stearyl amine salts.   |    |
| <b>4</b> 5 | Example 2.  Gelled non-polar organic liquids containing 1% by wt. of N°, N°-dicapryloylornithine, N-lauroylglutamic acid-a, y-di-n-butylamide or stearylamine N°, N°-dilauroyllysinate were prepared by the procedure set out in Example 1,  | 45 |
| 50         | but with the difference that the gelling agents were disolved in the non-polar organic liquids at a temperature of 75—120°C. The shear strengths of the gels obtained with a wide variety of non-polar organic liquids are set out in Table 2.   | 50 |

TABLE 2

|   | Shear strength of gel                           |   |   |
|---|---|---|---|
| Non-polar<br>organic liquid                       | Na,Nô-di-<br>capryloyl-<br>ornithine<br>(g/cm²) | N-lauroylglutamic<br>acid-a,y-<br>di-n-butylamide<br>(g 'cm²) | Stearylamine<br>Na,Ne-dilauroyl-<br>lysinate<br>(g'cm²) |
| Benzene   | 30  | 131   | 12 -  |
| Cyclohexane                                       | 14  | 75  | 18  |
| Heavy oil *1                                      | 11  | 202   | 19  |
| Spindle oil *2                                    | 17  | 345   | 30  |
| Turbine oil *3                                    | 21  | 177   | 32  |
| Carbon tetrachloride                              | 102   | 10  | 12  |
| Dioctyl sebacate                                  | 25  | 264   | 45  |
| n-Butyl acetate                                   | 11  | 13  | 12  |
| Dioctyl phthalate                                 | 15  | 87  | 33  |
| Methyl isobutyl ketone                            | 2.5   | 26  | 3.8   |
| Liquid paraffin                                   | 16  | 398   | 57  |
| Polyoxypropylene *4<br>glycol mono<br>butyl ether | 22  | . 173   | 80  |
| Polyoxyethylene *5 glycol mono oleate             | 23  | 168   | 75  |
| Corn oil  | 10  | 125   | 89  |
| Rape seed oil                                     | 12  | 189   | 108   |
| Soya bean oil                                     | 13  | 353   | 122   |
| Tricresyl phosphate                               | 2.1   | 217   | 15  |
| Gasoline  | 12  | 162   | 16  |
| Gas oil   | 15  | 211   | 19  |
| Xylene  | 24  | 85  | 13  |
| Toluene   | 16  | 62  | y ·   |
| Cotton seed oil                                   | 11  | 174   | 103   |
| Diisobutylketone                                  | 3.0   | 35  | 4.5   |
| Diethyl phthalate                                 | 15  | 83  | 28  |
| Octane  | 8.0   | 63  | 15  |

10

Footnotes to Table 2.

- \*1: Viscosity of 2.1 C.P. (50°C)
- \*2: Viscosity of 4.0 cst (50°C) and pour point below -5°C
- \*3: Viscosity of 17.5 22.5 cst (50°C) and pour point below -5°C
- \*4: Contained 40 oxypropylene units
- \*5: Contained 5 oxyethylene units

Example 3. Gelled kerosene containing different amounts of N-lauroyl-glutamic acid-a,ydi-n-octylamide was prepared in the manner described in Example 1. The results of shear strength tests carried out on the gels obtained are set out in Table 3.

TABLE 3

| Gelling agent (wt. %) | Shear strength of kerosene gel (g/cm²) |  |
|-----------------------|--|--|
| 0.1                   | 6                                      |  |
| 0.5                   | 56                                     |  |
| 1.0                   | 130                                    |  |
| 2.0                   | 387                                    |  |
| 5.0                   | 475                                    |  |

Example 4.

Three grams of the N-acylamino acid derivatives shown in Table 4, the derivatives being in powder form, were admixed in 100 grams of a non-polar organic liquid.

The mixture obtained in each case was vigorously shaken for 10 minutes and allowed to stand overnight. In the morning, the mixtures were found to be gelled. Shear tests carried out on the gels gave the results set out in Table 4.

TABLE 4

| Derivative of N-acylamino acid                        | Non-pólar organic<br>liquid | Shear<br>strength of<br>gel (g/cm²) |
|---|-----------------------------|-------------------------------------|
| N <sup>α</sup> , N <sup>δ</sup> -dicapryloylornithine | Benzene                     | 45                                  |
| ,,  | . Carbon tetrachloride      | 140                                 |
| ,,  | Kerosene                    | 23                                  |
| N-lauroylglutamic acid-<br>α,y-di-n-octylamide        | Benzene                     | 85                                  |
| N-lauroylglutamic acid-<br>α,γ-di-n-stearylamide      | Benzene                     | 4.5                                 |

5

10

BNSDOCID: <GB\_\_\_1485694A\_\_I\_>

5

10

Example 5.

A series of experiments was carried out using N-lauroyl-glutamic acid- $\alpha$ , din-butylamide as gelling agent for various non-polar organic liquids. In each experiment, one gram of N-lauroylglutamic acid- $\alpha$ , din-butylamide was dissolved in 5 ml of ethyl acetate at a temperature of 60°C with stirring. This solution was added to 100 grams of the selected non-polar organic liquid with stirring. After standing for one hour, the solution was gelled. The results of shear strength tests carried out on the gels obtained are set out in Table 5.

TABLE 5

| Non-polar organic liquid | Shear strength of gel (g/cm²) |
|--------------------------|-------------------------------|
| Ligroin                  | 89                            |
| Kerosene                 | 185                           |
| Gasoline                 | 83                            |
| Heavy oil                | 145                           |
| Liquid paraffin          | 298                           |
| Olive oil                | 176                           |
| Soya bean oil            | 212                           |
| Rape seed oil            | 153                           |
| Corn oil                 | 97                            |

Example 6.

Gels of non-polar organic liquids were prepared containing 1° by wt. of N°, N'-dicapryloyllysine laurylester as gelling agent in the manner described in Example 5. The results of shear strength tests carried out on the gels are set out in Table 6.

TABLE 6

| Non-polar organic liquid                     | Shear strength of gel<br>(g'cm²) |
|--|----------------------------------|
| Kerosene                                     | 18                               |
| Cyclohexane                                  | 13                               |
| Heavy oil                                    | 25                               |
| Liquid paraffin                              | 31                               |
| Dioctyl phthalate                            | 16                               |
| Polyoxypropylene-<br>glycol_monobutylether*1 | 52                               |
| Olive oil                                    | 59                               |

<sup>\*1:</sup> Containing 40 oxypropylene units

10

15

20

5

10

Example 7.

A series of experiments was carried out using N-lauroylglutamic acid as gelling agent. In each experiment 4 grams of N-lauroylglutamic acid were dissolved in 10 ml of acetone at a temperature of 55°C. This solution was added to 100 grams of a non-polar organic liquid with stirring.

After standing overnight, the mixture had become gelled. The results of tests on shear strength of the gel obtained are set out in Table 7.

TABLE 7

| Non-polar organic liquid | Shear strength of gel<br>(g.'cm²) |
|--------------------------|-----------------------------------|
| Benzene                  | 3.0                               |
| Spindle                  | 4.5                               |
| Olive oil                | 3.8                               |

Example 8.

Further experiments of the type reported in Example 7 were carried out using N-lauroylalanine as gelling agent. The results of shear strength tests on the gels obtained are set out in Table 8.

TABLE 8

| Non-polar organic liquid | Shear strength of gel (g/cm²) |
|--------------------------|-------------------------------|
| Kerosene                 | 28                            |
| Heavy oil                | 17                            |
| Spindle oil              | 29                            |
| Turbine oil              | 32                            |

Example 9.

One gram samples of each of the N-acylamino acid derivatives shown in Table 9 were each admixed with 90 grams of a non-polar organic liquid as shown in Table 9 and the mixture was heated at a temperature of 80°C for 10 minutes.

10 grams of water were added to the solution obtained and the mixture obtained was vigorously shaken at a temperature of 80°C.

After allowing the suspension obtained to cool to room temperature, the suspension underwent gelling. The results of the shear strength tests carried out on the various gels are set out in Table 9.

10

15

20

5

10

15

20

TABLE 9

| Derivative of N-acylamino<br>acid                                 | Non-polar organic<br>liquid                      | Shear strength<br>of gel<br>(g/cm²) |
|---|--|-------------------------------------|
| N-lauroylglutamic acid-α,γ-<br>dis tearylamide                    | Kerosene   | 15                                  |
| ,,,   | Heavy oil  | 21                                  |
| ,,,   | Liquid paraffin                                  | 109                                 |
| **  | Polyoxypropylene<br>glycol monobutyl<br>ether *1 | 20                                  |
|   | Olive oil  | 91                                  |
| N-lauroylvaline lauryl-amide                                      | Liquid paraffin                                  | 3.5                                 |
| N <sup>a</sup> ,N <sup>c</sup> -dicapryloyllysine<br>lauryl ester | ,, ,,  | 15                                  |
| Stearylamine Na,N- dicapryloyllysinate                            | Polyoxypropylene<br>glycol monobutyl<br>ether *1 | 31                                  |
| N-Capryloylleucine<br>laurylamide                                 | Liquid paraffin                                  | 8.3                                 |
| Laurylamine N-lauroyl-<br>phenylalaninate                         | Olive oil  | 62                                  |

\*1: Containing 40 oxypropylene units

Example 10.

One gram of N-lauroylglutamic acid- $\alpha, p$ -di-n-butyramide was dissolved in 5 ml of methanol at a temperature of 50°C. The solution obtained was added slowly to a mixture of 70 grams of olive oil and 30 grams of water with vigorous stirring. After allowing the suspension to stand for 20 minutes it underwent gelling. The shear strength of the gel was 189 g/cm<sup>2</sup>.

Example 11.

(a) 0.5 gram of N-lauroylglutamic acid- $\alpha$ , y-di-n-octylamide was dissolved in 5 ml of benzene at a temperature of 50°C. This solution was added to 1 litre of sea water in which were suspended 25 grams of heavy oil and the water was stirred for 30 seconds. After standing for 20 minutes, the heavy oil solidified and floated to the surface of the sea water.

The mixture obtained was then filtered through a wire gauze of 0.05" mesh and 31 grams of solidified heavy oil were collected. The oil contained benzene and a small amount of water. The amount of heavy oil in the filtrate was found to be below 75 ppm by measurement according to Japanese Industrial Standards (JIS) K-0102.

(b) A similar experiment was carried out using crude oil (Iranian heavy) in place of the heavy oil.

Thirty grams of crude oil were collected and the content of crude oil in the filtrate obtained at the end of the experiment was below 100 ppm.

| 13 | 1,485,694   | 13 |
|----|---|----|
| 5  | Example 12.  0.5 ml or 0.1 ml samples of ethanol containing 10 g/dl of a derivative of an Nacylamino acid as specified in Table 10 were each added to 5 l of water in which 0.5 to 2.5 grams of a non-polar organic liquid, as specified in Table 10, were suspended. The mixtures obtained were stirred vigorously for 10 minutes and then allowed to stand for one hour.  The non-polar organic liquid solidified in each case and floated to the surface | 5  |
| 10 | of the water. The solidified liquid was removed by filtration.  The residual amount of non-polar organic liquid in the filtrate was measured in each case by the methods of JIS K—0102. The amounts determined are set out  | 10 |

## TABLE 10

|   | Admixed<br>Volume of        |                   | Organic liquid<br>content (ppm) |                    |
|---|-----------------------------|-------------------|---------------------------------|--------------------|
| Derivative of<br>N-acylamino acid               | ethanol<br>solution<br>(ml) | Organic<br>liquid | In initial suspension           | In the<br>filtrate |
| N-lauroylglutamic acid-a,y-<br>di-n-butyl amide | 0.5                         | Heavy oil         | 100                             | 0.57               |
| N-caproylglutamic acid-a,y-<br>di-stearylamide  | ٠,                          | ••                | ,,                              | 0.71               |
| N-lauroyl valine laurylamide                    | ,,                          | ,,                | ,,                              | 0.63               |
| N-palmitoyl methionine<br>n-butyl amide         | ,,                          | ,,                | ••                              | 3.95               |
| N-stearoylleucine benzyl amide                  | ,,                          | **                | 73                              | 3.55               |
| N-lauroylphenylalanine<br>lauryl amide          | ,,                          | **                | ,,                              | 3.50               |
| N-capryloylglycine stearyl amide                | ,,                          | **                | . 22                            | 4.36               |
| N-stearoyl serine 2-ethyl<br>hexyl amide        | ,,                          | ,,                | 17                              | 5.93               |
| Nª,N&dicapryloyllysine<br>lauryl ester          | ,,                          | **                | "                               | 0.59               |
| N-lauroylamino caroic<br>acid lauryl amide      | τ.                          | ••                | **                              | 3.83               |
| Stearylamine Na,Ne-di-<br>capryloyllysinate     | ,,                          | ••                | ••                              | 0.90               |
| N-lauroylglutamic acid-α,y-<br>di-n-butyl amide | ,,                          | Soya bean<br>oil  | ,,                              | 0.36               |
| "   | ,,                          | Turbine<br>oil    | ,,                              | 0.42               |
| N-lauroylglutamic acid-α,y-<br>di-stearylamide  | 0.1                         | Heavy oil         | ,,                              | 3.1                |
| ,,  | 0.5                         | . ,,              | - 500                           | 2.9                |
| ,,  | 0.1                         | Soya bean<br>oil  | 100                             | 2,6                |
| . "   | 0.5                         | **                | 500                             | 3.9                |

Example 13.

A series of experiments was carried out in the same manner as described in Example 12 using water in which the selected non-polar organic liquid was emulsified with a surfactant. The results of the experiments are set out in Table 11. In each case the surfactant used was sodium dodecyl sulphate employed in the water in an amount of 100 ppm.

5

5

15

20

30

5

15

20

25

30

TABLE 11

|   | Admixed<br>volume of<br>ethanol | Non-polar         | Content of non-polar organic liquid (ppm) |                 |
|---|---------------------------------|-------------------|---|-----------------|
| Derivative of<br>N-acyl amino acid              | solution<br>(ml)                | organic<br>liquid | In initial suspension                     | In the filtrate |
| N-lauroylglutamic acid-α-γ-<br>di-n-butyl amide | 0.5                             | Soya bean<br>oil  | 100                                       | 1.5             |
| ***   | ,,                              | Heavy oil         | ,,  | 3.8             |
| N-lauroylglutamic acid-a,y-<br>di-stearylamide  | >7                              | ,,                | ,,  | 2.5             |
| ,,  | ,,                              | Turbine<br>oil    | "   | 2.1             |

Example 14.

One gram of N-lauroylglutamic acid- $\alpha$ , p-di-n-butyramide was dissolved in 45 grams of liquid paraffin at a temperature of 120°C with stirring. 48.8 grams of olive oil, 5 grams of white petroleum jelly, 0.2 grams of rose essence and a very small quantity of colouring matter were added to the liquid paraffin solution with stirring at a temperature of 80°C.

The resultant solution was cooled to room temperature. After standing for one hour, the solution gelled to form a clear hair conditioner.

10 Example 15. 10

One gram of N°, N°-dicapryloyl lysine lauryl ester was dissolved at a temperature of 90°C in 68.8 grams of a solution constituted by 54.1 grams of liquid paraffin in which 4 grams of squalene, 4 grams of isopropylmyristate, 2 grams of beeswax, 4.5 grams of polyoxyethylene stearyl ether and 0.2 grams of methyl benzoate were dissolved. 27 grams of water and 3 grams of polyethylene glycol were added to this solution and the suspension obtained was shaken vigorously at a temperature of 80°C to form an emulsion. temperature of 80°C to form an emulsion.

The emulsion was then cooled to a temperature of 60°C and 0.3 grams of rose essence were added thereto with stirring. The emulsion was then cooled to room temperature and allowed to stand overnight. The emulsion gelled. The gel

obtained was a cleansing-jelly which had good spreading properties and was situated for application to the skin.

Example 16. Forty-five grams of chlorinated rubber (20% by wt. solution in toluene; viscosity of 10 centipoise), 18 grams of chlorinated paraffin, 58 grams of titanium dioxide, 70 grams of toluene, 4 grams of xylene and 4 grams of butyl acetate were thoroughly mixed in a ball mill to form a paint. 25

Six grams of hot toluene containing 1 gram of N-lauroylglutamic acid- $\alpha$ , $\gamma$ -din-butyramide dissolved therein were added with stirring to the paint which was allowed to stand overnight. The mixture obtained underwent gelling

The thixotropic properties of this paint (1) and of a similar paint (2) containing no N-lauoylglutamic acid- $\alpha$ ,p-di-n-butyramide as a control were measured with a viscometer, type B8L (a product of Tokyo Keiki Co., Japan) and are set out in Table 12.

BNSDOCID: <GB\_\_\_1485694A\_\_I\_>

10

15

20

25

30

5

15

30

#### TABLE 12

|     | (A) Viscosity at 6 rpm (cp) | (B) Viscosity at 60 rpm<br>(cp) | Ratio<br>A/B |
|-----|-----------------------------|---------------------------------|--------------|
| (1) | 8400                        | 2400                            | 3.5          |
| (2) | 550                         | 540                             | 1.0          |

WHAT WE CLAIM IS:-

1. A process for preparing a gel which comprises (A) homogeneously admixing a non-polar organic liquid having a dielectric constant of not more than 20 and having a solubility of not more than 10% by weight in water, with at least one N-acylamino acid or derivative thereof employed in an amount sufficient to gel the non-polar organic liquid, N-acylamino acid or derivative thereof possessing the general formula:

wherein RCO is an aliphatic acyl radical containing from 2 to 30 carbon atoms 10 or an aralkyl acyl radical; Z is a group of formula -OH, -OR1, -NH2, -NHR2,

or  $-O^-R_5NH_3^+$ , wherein  $R_1$ ,  $R_2$ ,  $R_3$ ,  $R_4$  and  $R_5$  are aliphatic hydrocarbyl radicals containing from 1 to 30 carbon atoms or aralkyl radicals; X is hydrogen or a methyl radical, and Y is a group of formula

wherein R<sub>6</sub> is an alkyl radical containing from 1 to 4 carbon atoms, a benzyl radical, a phenyl radical, or a radical of formula -CH2OH,

-CH<sub>2</sub>SH, -CH<sub>2</sub>CH<sub>2</sub>S-CH<sub>3</sub>, -(CH<sub>2</sub>)<sub>1</sub>-COZ, wherein 1 is 1 or 2 and Z has the aforesaid meaning, -(CH<sub>2</sub>)<sub>m</sub>NHCOR wherein m is 3 or 4 and RCO 20 is as above, or  $-(CH_2)_n$ , wherein n is an integer of from 1 to 6, with the proviso that when Z is  $-OR_1$ , Y is

wherein  $R_6$  is —  $(CH_2)_m$ NHCOR and that at least one or R, R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub> and R<sub>5</sub> is an aliphatic hydrocarbyl radical containing from 5 to 30 carbon atoms. 25 and (B) allowing the mixture to stand until gel formation occurs.

2. A process as set forth in Claim 1, wherein the said N-acylamino acid or

derivative thereof is added to said non-polar organic liquid in an amount of from 0.05% to 20% by weight of the non-polar organic liquid.

3. A process as set forth in Claim 2, wherein the said N-acylamino acid or

BNSDOCID: <GB\_\_\_1485694A\_\_,I\_>

| 17  | 1,485,694  | 17  |
|-----|--|-----|
|     | derivative thereof is added to said non-polar organic liquid in an amount of from 0.01% to 10% by weight of the non-polar organic liquid.  |     |
|     | 4. A process as set forth in Claim 1, wherein the homogeneous admixing is  |     |
| 5   | carried out at temperatures ranging from room temperature up to the boiling point  |     |
| J   | of said non-polar organic liquid.  | 5   |
|     | 5. A process as set forth in Claim 1 or 2, wherein said N-acylamio acid or derivative thereof is admixed with said non-polar organic liquid in the form of very  |     |
|     | the powdered particles of as a solution.   |     |
|     | 6. A process as set forth in any one of the preceding claims, comprising the   |     |
| 10  | additional steps of (C) dissolving the N-acylamino acid or derivative in the non   | 10  |
|     | polar organic solvent at an elevated temperature and (D) cooling the resultant   |     |
|     | solution to a temperature at which gel formation occurs on standing.  7. A process as set forth in any one of the preceding claims, wherein said non-  |     |
|     | polar organic riquid is a righted hydrocarbon.   |     |
| 15  | 8. A process as set forth in Claim 7 wherein said liquid hydrogarbon is  | 1.5 |
|     | gasonile, a naprina, kerosene, gas oil, heavy oil, crude oil spindle oil turbine oil   | 15  |
|     | inquid parairin, benzene, xviene, toluene, nexane, hentane, octane or cycloberone  |     |
|     | 9. A process as set forth in any one of Claims 1 to 6, wherein said non-polar organic liquid is a fatty oil.   |     |
| 20  | 10. A process as set forth in Claim 9, wherein said fatty oil is corn oil sour   | 20  |
|     | bean oil, olive oil, rape seed oil, cotton seed oil, sardine oil herring oil or whole oil  | 20  |
|     | 11. A process as set forth in any one of Claims 1 to 6 wherein said non polar  |     |
|     | organic riquid is a sincone oil.   |     |
| 25  | 12. A process as set forth in any one of Claims 1 to 6, wherein said non-polar organic liquid is an ester.   |     |
|     | 13. A process as set forth in Claim 12, wherein said ester is but yell acetate amy   | 25  |
|     | acctate, diethyl phinalate, dibutyl phinalate, dioctyl phinalate, diethyl sebacate   |     |
|     | diocity seducate, though phosphate or tricresvi phosphate  |     |
| 30  | 14. A process as set forth in any one of Claims 1 to 6, wherein said non-polar organic liquid is an aldehyde or ketone.  |     |
|     | 15. A process as set forth in Claim 14, wherein the non-polar organic liquid is  | 30  |
|     | methyl isobutyl ketone, diisobutyl ketone or anisaldehyde  |     |
|     | 16. A process as set forth in any one of claims 1 to 6, wherein said non-polar   |     |
| 35  | organic liquid is a liquid polyoxyalkylene monalkyl ether.   |     |
|     | 17. A process as set forth in Claim 16, wherein said liquid polyoxyalkylene monoalkyl ether is a liquid polyoxyethylene mono stearyl ether, a polyoxy-   | 35  |
|     | ctrylene incholauryl ether containing 4 to 6 oxyethylene units or a polyeyr  |     |
|     | propylene monoalkyl ether wherein the alkyl radical has from 4 to 12 carbon  |     |
| 40  | atoms and contains to to be oxypropylene units   | •   |
|     | 18. A process as set forth in Claim 17, wherein said liquid polyoxyalkylene mono alkyl ether is a liquid polyoxypropylene monobutyl ether, liquid polyoxypropylene monobutyl ether, liquid polyoxypropylene monobutyl ether, liquid polyoxypropylene monobutyl ether, liquid polyoxypropylene monobutyl ether. | 40  |
|     | propyrene monostearyl ether or liquid polyoxynronylene monostearyl ether   |     |
|     | 17. A process as set torth in any one of Claims 1 to 6 wherein said non notes  |     |
| 45  | organic injuly is a liquid polyoxyalkylene glycol tatty acid ester   |     |
| 7.5 | 20. A process as set forth in Claim 19 wherein said polyoxyalkylene glycol fatty acid ester is polyoxyethylene glycol  | 45  |
|     | fatty acid ester is polyoxyethylene glycol lauric or oleic acid ester containing from 4 to 6 oxyethylene units.  |     |
|     | 21. A process as set forth in any one of Claims 1 to 6, wherein the non pole-  |     |
| 50  | organic inquid is a chiorinated hydrocarbon.   |     |
| 50  | 22. A process as set forth in Claim 21, wherein the chlorinated hydrocarbon is carbon tetrachloride, tetrachloroethylene or chlorobenzene.   | 50  |
|     | 23. A process as set forth in any one of the process.  |     |
|     | 23. A process as set forth in any one of the preceding claims which is carried out using a said N-acylamino acid possessing the general formula  |     |
|     | RCO-N-Y-C-OH   |     |
|     | H O  |     |
|     |  |     |
| 55  | wherein RCO is an aliphatic acyl radical containing from 5 to 30 carbon atoms and  | 55  |
|     | Y is a group of formula  | 23  |
|     | —СН—,  |     |
|     | <del></del> ,  |     |

15

20

25

30

35

5

10

20

25

wherein  $R_{\epsilon}$  is an alkyl radical containing from 1 to 4 carbon atoms or a radical -(CH<sub>2</sub>)<sub>m</sub>NHCOR wherein m is 3 or 4 and RCO is the same as above.

24. A process as set forth in Claim 23, wherein said N-acylamino acid is N-lauroyl-α-alanine, Nα, Nδ-dicaproyl ornithine, Nα, Nδ-didecanoyl ornithine, Nα, Nδdicapryloyl ornithine, N°, N<sup>8</sup>-dilauroylornithine, N°, N<sup>8</sup>-dicocoylornithine, N°, N′-dicaproyllysine, N°, N′-dicapryloyllysine, N-lauroyl valine or N-lauroyl glutamic

25. A process as claimed in any one of Claims 1 to 22 which is carried out in the presence of an N-acylamino derivative having the general formula

wherein RCO and R, have the meanings set out in Claim 1 and Y is a group of formula

where  $R_6$  is  $-(CH_2)_mNHCOR$  wherein m is 3 or 4 and RCO has the aforesaid meaning with the proviso that at least one of R and  $R_1$  is an aliphatic hydrocarbon 15

meaning with the proviso that at least one of R and R<sub>1</sub> is an aliphatic hydrocarbon radical containing from 5 to 30 carbon atoms.

26. A process as set forth in Claim 25, wherein said N-acylamino acid derivative is a N<sup>a</sup>,N<sup>8</sup>-dicapryloylornithine alkyl ester, N<sup>a</sup>,N<sup>8</sup>-dilauroylornithine alkyl ester, N<sup>a</sup>,N<sup>8</sup>-distearoylornithine alkyl ester, N<sup>a</sup>,N<sup>8</sup>-dicapryloyllysine alkyl ester, N<sup>a</sup>,N<sup>8</sup>-distearoyllysine alkyl ester or N<sup>a</sup>,N<sup>8</sup>-ditallowyllysine alkyl ester.

27. A process as claimed in Claim 26, wherein the alkyl esterifying group of the alkyl ester contains from 6 to 18 carbon atoms.

the alkyl ester contains from 6 to 18 carbon atoms.

28. A process as set forth in any one of Claims 1 to 22, which is carried out in the presence of a N-acylamino acid derivative having the general formula

wherein RCO has the general meaning set out in Claim 1, Z is -NH2, -NHR2 or

$$-N$$
 $R_3$ ,

wherein R<sub>2</sub>, R<sub>3</sub> and R<sub>4</sub> have the general meanings set out in Claim 1, and Y is a 30 group of formula

wherein  $R_6$  is an alkyl radical containing from 1 to 4 carbon atoms, a benzyl radical, or a radical of formula — $CH_2CH_2$ —S— $CH_3$ , — $(CH_2)_1$ —COZ, wherein I is 1 or 2 and Z has the aforesaid meaning, — $(CH_2)_m$ NHCOR, wherein m is 3 or 4 and RCO has the aforesaid meaning and — $(CH_2)_m$ —A wherein n is an integer of 1 to 6 and A 35

10

15

20

25

30

35

40

45

50

55

5

25

30

35

40

45

50

55

with the proviso that at least one of R, R2, R3 and R4 is an aliphatic hydrocarbon radical containing from 5 to 30 carbon atoms.

29. A process as set forth in Claim 28, wherein said N-acylamino acid related compound is a N-acetyl glutamic acid- $\alpha$ ,p-dialkyl amide wherein the alkyl radicals have 12 to 18 carbon atoms, N-capryloylglutamic acid- $\alpha$ ,p-diamine, a N-capryloylglutamic acid- $\alpha$ ,p-dialkylamide wherein the alkyl radicals have 1 to 18 carbon atoms, N-lauroylglutamic acid- $\alpha, \gamma$ -diamide, a N-lauroylglutamic acid- $\alpha, \gamma$ dialkylamide wherein the alkyl radicals have 1 to 18 carbon atoms, N-cocoylglutamic acid- $\alpha$ , $\gamma$ -dialkylamide, a N-cocoylglutamic acid- $\alpha$ , $\gamma$ -dialkylamide wherein the alkyl radicals have 1 to 18 carbon atoms, N-stearoylglutamic acid- $\alpha$ , $\gamma$ -dialkylamide wherein the alkyl radicals have 1 to 18 carbon atoms, N-stearoylglutamic acid- $\alpha$ , $\gamma$ -10 diamide, a N-stearoylglutamic acid- $\alpha$ , $\gamma$ -dialkylamide wherein the alkyl radicals have 1 to 18 carbon atoms, N-(tallowyl)glutamic acid- $\alpha$ , $\gamma$ -diamide, a N-(tallowyl)glutamic acid- $\alpha$ ,p-dialkylamide wherein the alkyl radicals have 1 to 18 carbon atoms, a N°,N'-diacetyllysine alkylamide wherein the alkyl radical has 8 to 18 carbon atoms, Na, Ne-dicapryloyllysine amide, a Na, Ne-dicapryloyllysine alkyl-15 amide wherein the alkyl radical has 1 to 18 carbon atoms, N°, N'-dilauroyllysine amide, a N°, N'-dilauroyllysine alkylamide wherein the alkyl radical has 1 to 18 carbon atoms, N°, N'-dicocoyllysine alkylamide wherein the alkyl radical has 1 to 18 carbon atoms, N°, N'-dicocoyllysine amide, a N°, N'-dicocoyllysine alkylamide wherein the alkyl radical has 1 to 18 carbon atoms, N°, N'-distearoyllysine amide, a N°,N'-distearoyllysine alkylamide, wherein the alkyl radical has I to 18 carbon atoms, N°,N'-di(tallowyl)lysine amide or a N°,N'-di(tallowyl)lysine alkylamide 20 wherein the alkyl radical has 1 to 18 carbon atoms.

30. A process as set forth in any one of Claims 1 to 22, which is carried out in the presence of a N-acylamino acid derivative having the general formula

wherein RCO and R, have the meanings set out in Claim 1 and Y is a group of formula

wherein  $R_6$  is an alkyl radical containing from 1 to 4 carbon atoms, a benzyl radical or a radical of formula  $-(CH_2)_mNHCOR$  wherein m is 3 or 4 and RCO has the aforesaid meaning with the proviso that at least one of R and  $R_5$  is an aliphatic hydrocarbyl radical containing from 5 to 30 carbon atoms.

31. A process as set forth in Claim 30, wherein said N-acylamine acid relative

compound is a N°, N'-dicaproyllysine alkylamine salt wherein the alkylamine contains from 4 to 18 carbon atoms, a N°, N'-dilauroyllysine alkylamine salt wherein the alkylamine has 4 to 18 carbon atoms, a No, No-dicocoyllysine alkylamine salt wherein the alkylamine has 4 to 18 carbon atoms, a Na, Na distearoyllysine alkylamine salt wherein the alkylamine has 4 to 18 carbon atoms, a Na, Nadi(tallowyl or hydrogen tallowyl)lysine alkylamine salt wherein the alkylamine has 4 to 18 carbon atoms, a N°,N<sup>5</sup>-dicapryloylornithine alkylamine salt wherein the alkylamine has 8 to 18 carbon atoms, a N°, N°-dilauroylornithine alkylamine salt wherein the alkylamine has 4 to 18 carbon atoms, a N°, N°-dicocoylornithine alkylamine salt wherein the alkylamine has 4 to 18 carbon atoms, a Na, No-distearoylornithine alkylamine salt wherein the alkylamine has 4 to 18 carbon atoms, a Na, No. ditallowylornithine alkylamine salt wherein the alkylamine has 4 to 18 carbon atoms, a N-lauroylphenylalanine alkylamine salt wherein the alkylamine has 8 to 18 carbon atoms, a N-cocoylphenylalanine alkylamine salt wherein the alkylamine has 8 to 18 carbon atoms, a N-stearoylphenylalanine alkylamine salt wherein the alkylamine has 8 to 18 carbon atoms or a N-tallowylphenylalanine alkylamine salt wherein the alkylamine has 8 to 18 carbon atoms.

32. A process for the production of a gel as claimed in claim 1 substantially as described in any one of the experiments set out in the foreging Examples 1 to 16. 33. A gel which comprises a homogeneous mixture of (a) a non-polar organic

liquid having a dielectric constant of not more than 20 and having a solubility in water of not more than 10% by weight and (b), present in an amount sufficient to gel the non-polar organic liquid, at least one N-acylamino acid or derivative thereof having the general formula:

20

25

30

35

5

10

15

20

25

40

wherein RCO, X, Y and Z have the meanings set out in Claim 1.

34. A gel as set forth in Claim 33, which contains the at least one N-acylamino acid or derivative thereof in an amount of from 0.05 to 20%, by weight of the nonpolar organic liquid.

35. A gel as set forth in Claim 34, which contains the at least one N-acylamino acid or derivative thereof in an amount of from 0.1 to 10°, by weight of the non-

polar organic liquid. 36. A gel as set forth in any one of Claims 33 to 35, wherein the non-polar organic liquid is a petroleum hydrocarbon, a lubricating oil, a pure hydrocarbon. an ester, a ketone, an aldehyde, a chlorinated hydrocarbon, a phosphoric ester, a liquid polyoxyalkylene monoalkyl ether or a liquid polyoxyalkylene glycol fatty acid ester, or a mixture of two or more such liquids.

37. A gel as set forth in Claim 36, wherein the non-polar organic liquid is

gasoline, naphtha, kerosene, gas oil, heavy oil, crude oil, spindle oil, turbine oil, liquid paraffin, benzene, xylene, toluene, hexane, heptane, octane, cyclohexane, corn oil, soya bean oil, olive oil, rape seed oil, cotton seed oil, butyl acetate, diethyl phthalate, dioctyl phthalate, dioctyl sebacate, tricresyl phosphate, methyl isobutyl ketone, diisobutyl ketone, liquid polyoxyethylene monolauryl ether, liquid polyoxyethylene monostearyl ether, liquid polyoxypropylene monobutyl ether, liquid polyoxypropylene monolauryl ether, liquid polyoxypropylene monostearyl ether, liquid polyoxyethylene glycol lauric acid ester, liquid polyoxypropylene glycol oleic acid ester or a mixture of two or more said liquids.

38. A gel as set forth in any one of Claims 33 to 37, which contains a said N-

acylamino acid which possesses the general formula:

wherein RCO is an aliphatic acyl radical containing from 5 to 30 carbon atoms and Y is a group of formula

wherein R6 is an alkyl radical containing from 1 to 4 carbon atoms or a radical 30 (CH<sub>2</sub>)<sub>m</sub>NHCOR wherein m is 3 or 4 and RCO is the same as the above.

39. A gel as set forth in any one of Claims 33 to 37, which contains a said Nacylamino acid derivative having the general formula:

wherein RCO and R<sub>1</sub> have the meanings set out in Claim 1 and Y is a group of 35 formula

where R<sub>6</sub> is -(CH<sub>2</sub>)<sub>m</sub>NHCOR wherein m is 3 or 4 and RCO has the aforesaid meaning, with the proviso that at least one of R and R, is an aliphatic hydrocarbon

radical containing from 5 to 30 carbon atoms.

40. A gel as set forth in any one of Claims 33 to 37, which contains a said Nacylamino acid derivative having the general formula:

wherein RCO has the general meaning set out in Claim 1, Z is -NH2, -NHR2 or

$$-N$$
 $R_4$ ,

wherein R2, R3 and R4 have the general meanings set out in Claim 1, and Y is a group of formula

5

10

15

5

wherein  $R_6$  is an alkyl radical containing from 1 to 4 carbon atoms, a benzyl radical or a radical of formula — $CH_2CH_2$ —S— $CH_3$ , — $(CH_2)_1$ —COZ, wherein 1 is 1 or 2 and Z has the aforesaid meaning, — $(CH_2)_mNHCOR$ , wherein m is 3 or 4 and RCO has the aforesaid meaning and — $(CH_2)_m$ —A wherein n is an integer of 1 to 6 and A in

10

with the proviso that at least one of R, R<sub>2</sub>, R<sub>3</sub> and R<sub>4</sub> is an aliphatic hydrocarbon

radical containing from 5 to 30 carbon atoms.

41. A gel as set forth in any one of Claims 33 to 37, which contains a said Nacylamino acid derivative having the general formula:

15

$$RCO - N - Y - C - O^-R_5NH_3^+ H O$$

wherein RCO and R, have the meanings set out in Claim 1 and Y is a group of formula

—CH— R<sub>6</sub>

20

25

wherein  $R_6$  is an alkyl radical containing from 1 to 4 carbon atoms, a benzyl radical or a radical of formula —  $(CH_2)_mNHCOR$  wherein m is 3 or 4 and RCO has the aforesaid meaning with the proviso that at least one of R and R, is an aliphatic hydrocarbyl radical containing from 5 to 30 carbon atoms.

42. A gel as claimed in Claim 33, substantially as described in any one of the experiments set out in the foregoing Examples 1 to 16.

HASELTINE, LAKE & CO..

20

25

Chartered Patent Agents, 28, Southampton Buildings, Chancery Lane, London, WC2A AT.,

> Temple Gate House, Temple Gate. Bristol, BS1 6PT., - and ---9 Park Square, Leeds, LSI 2LH. Agents for the Applicants.

Printed for Her Majesty's Stationery Office by the Courier Press, Learnington Spa, 1977. Published by the Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from which copies may be obtained.